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LETTER TO THE EDITOR

Antiferromagnetic coupling in Co/Ge superlattices

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Abstract. We have investigated interlayer coupling of Co/Ge superlattices. The present experiments obviously show that the coupling changes from ferromagnetic (F) to antiferromagnetic (AF) and finally to non-coupling (N) with the increase of Ge layer thickness. This coupling behaviour, as a function of the spacer thickness, is very similar to that of Fe/Si superlattices, although the coupling strength is much smaller than the latter: namely, ~ 0.05 erg cm^{-2} for Co/Ge and ~ 1.0 erg cm^{-2} for Fe/Si. Precise structural characterization indicates that diffused spacers at Co/Ge interfaces are responsible for the AF coupling. The same coupling behaviour has also been observed in Co/non-magnetic Co–Ge superlattices, where interdiffusion at the interfaces is entirely suppressed. All these results clearly demonstrate that the interlayer coupling between neighbouring Co layers is mediated by non-magnetic Co–Ge spacers.

Since the discovery of interlayer coupling in Fe/Si/Fe trilayers by Toscano *et al* [1], the Fe/Si superlattice has received much attention because of its unique coupling behaviour, which is quite different from the usual metal/metal superlattices (i.e., the coupling changes from ferromagnetic (F) to antiferromagnetic (AF) and then to non-coupling (N) with the increase of Si layer thickness up to around 20 Å) [2–4]. It has also been found that the coupling strength is very sensitive to the spacer composition [3, 4]. Therefore careful evaluation of interdiffusion is essential for quantitative discussion of the interlayer coupling behaviour. Although AF coupling in Fe/Si superlattices is well established experimentally, the mechanism for the coupling is not fully understood because of difficulties in characterization of very complex diffused spacer structures. If one could obtain more information on superlattices similar to Fe/Si, such as Fe/Ge, Co/Ge or Si, it would be very helpful for understanding the origin of the coupling behaviours. However, there are very few experiments on these remaining systems [5–8]. Briner *et al* [6] found AF coupling in Fe/Si/Fe but not in Fe/Ge/Fe, and speculated that the density of defect states in a spacer dominates the nature of the interlayer coupling. Later, the same group reported that very weak AF coupling can be induced in Fe/Ge/Fe when the sample is prepared and then annealed at certain limited conditions [7]. As mentioned above, most previous works have been performed for the superlattices using Fe as a ferromagnetic material, but not for the superlattices using Co.

In this letter we set out to demonstrate that AF coupling found in Fe/Si superlattices does indeed also occur in Co/Ge superlattices. Furthermore, based on the study of Co/Co–Ge superlattices, it is shown that a non-magnetic Co–Ge diffused spacer is responsible for the obvious AF coupling.

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A series of Co/Ge superlattices was grown on surface oxidized Si(100) substrates at ambient temperature in a dc magnetron sputtering system, with a base pressure lower than 6×10^{-7} Torr and argon pressure of 3 mTorr. The deposition rates for Co and Ge layers were $1.5 \sim 1.7$ Å s^{-1} and $1.8 \sim 2.0$ Å s^{-1}, respectively. The superlattices were grown with the Co layer thickness fixed at $t_{Co} = 30$ Å and the nominal Ge layer thickness t_{Ge} varied from 5–70 Å, with 22 bilayers. Another series of samples was also grown with the Ge layer thickness fixed at $t_{Ge} = 6, 10, 25$ Å and the nominal Co layer thicknesses t_{Co} varied from 10–50 Å. The periodic and crystallographic structures of these superlattices were characterized by both low- and high-angle x-ray diffraction (XRD) using Cu–Kα radiation. The superlattice period (Λ) was evaluated by the kinematical Bragg's law $\Lambda = l\lambda/2\sin\theta$ for lth order reflection. Here λ is the x-ray wavelength and θ is the observed diffraction angle. Since the lower order reflections are seriously influenced by the refraction effect, we first calculated Λ for all detected Bragg reflections and then accurately determined the saturation (true) period for larger l [9]. The crystalline coherence length (ξ) was estimated from the full width at half maximum (FWHM) of a Co(111) diffraction peak using Scherrer's relation [10]. Ferromagnetic resonance (FMR) measurements were also carried out with an electric paramagnetic resonance spectrometer in order to check the coupling states of the superlattices [3]. We define $J > 0$ for F coupling and $J < 0$ for AF coupling. The electric resistance of the samples was measured by a dc four-terminal method. Furthermore, we fabricated Co/Co_{1-x} Ge_x ($x = 0.42 - -0.81$) superlattices in order to investigate the material mediating the interlayer coupling between the adjacent Co layers.

Figure 1 shows the remanence ratio (M_r/M_s) and saturation field (H_s) of [Co(30 Å)/Ge(t_{Ge} Å)]$_{22}$ superlattices as functions of Ge layer thickness. It is clear that distinct AF coupling appears at $t_{Ge} \sim 10$ Å. The coupling constant J was evaluated by the following two methods. From magnetization measurements, the bilinear coupling constant J can be determined by $J = M_s H_s t_F/4$, where M_s is the saturation magnetization, H_s the saturation field and t_F the magnetic layer thickness. From FMR measurements, J can be evaluated by $J = (H^+ - H^-)M_s t_F$, where H^+ and H^- are the resonance fields for optical and accoustic modes, respectively. Both methods gave $J \sim -50$ merg cm^{-2}; this value being considerably smaller than that of Fe/Si ($J \sim -1.2$ erg cm^{-2}) [3]. Structural characterization of these layered structures was performed by the low- and high-angle XRD. The precisely determined superlattice period Λ_m is plotted as a function of Ge layer thickness, along with the designed

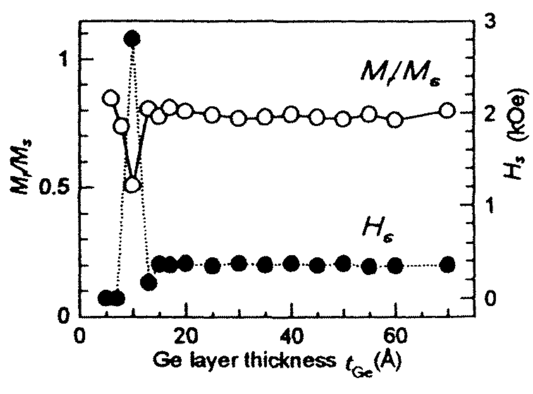

Figure 1. Dependence of the remanence ratio M_r/M_s (○) and the saturation field H_s (●) on the Ge layer thickness of [Co(30 Å)/Ge(t_{Ge} Å)]$_{22}$ superlattices.

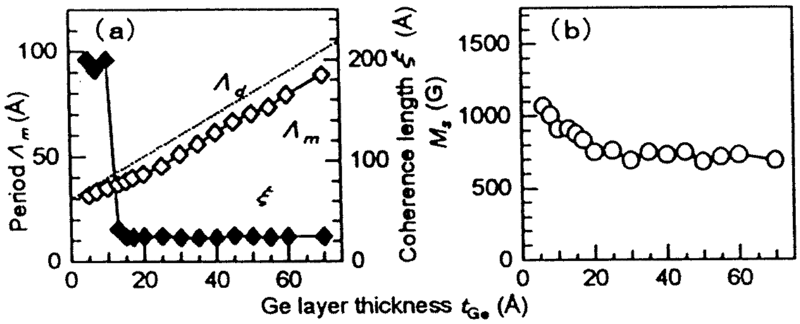

Figure 2. (a) Superlattice period Λ_m (◇) and coherence length ξ (◆) and (b) the saturation magnetization M_s (○) of [Co(30 Å)/Ge(t_{Ge} Å)]$_{22}$ superlattices as functions of Ge layer thickness. The dotted line in (a) shows the designed superlattice period Λ_d.

period (Λ_d) which is the simple summation of Co ($t_F = 30$ Å) and Ge layer thicknesses (t_{Ge}), as shown in figure 2(a). The increase of Λ_m is very gradual in the thickness range $t_{Ge} < 20$ Å, indicating severe interdiffusion at Co/Ge interfaces. In fact, an appreciable decrease of the saturation magnetization is observed in this layer thickness range, as shown in figure 2(b). These results clearly demonstrate that interdiffusion is extended up to $t_{Ge} = 20$ Å. The crystallinity of the diffused spacer was investigated by measuring the crystalline coherence length ξ of Co(111), as shown in figure 2(a). The spacer is crystalline for $t_{Ge} < 10$ Å and then abruptly changes to amorphous for larger thickness. All these structural analyses lead us to conclude that the spacer of Co/Ge superlattices mainly consists of the following three regions: diffused Co–Ge crystalline region for $0 < t_{Ge} \leqslant 10$ Å, diffused Co–Ge amorphous region for $10 < t_{Ge} \leqslant 20$ Å and amorphous Ge region for $t_{Ge} > 20$ Å. By comparing figures 1 and 2, we notice that all the interlayer coupling found in Co/Ge superlattices occurs within the thickness range of $t_{Ge} < 20$ Å. This means that the diffused Co–Ge layer is responsible for the interlayer coupling. It should be also noted that strong AF coupling found at $t_{Ge} = 10$ Å can be attributed to the crystalline Co–Ge diffused layer since coherence length ξ far exceeds each Co layer thickness.

Thus, we have observed obvious AF coupling in Co/Ge superlattices, and the coupling is mediated by the Co–Ge diffused layer. Direct evidence for AF coupling by Co–Ge is given by using non-magnetic Co–Ge alloy spacers instead of pure Ge, as described below. Table 1 lists electric resistivity ρ and saturation magnetization M_s for various kinds of Co_{1-x} Ge_x ($0.42 \leqslant x \leqslant 1.00$) single layers prepared in the present study. For all the compositions, they are amorphous and non-magnetic at room temperature, and the resistivity increases rapidly with Ge content x. By using these non-magnetic alloys, we prepared five series of [Co(30 Å)/Co_{1-x}

Table 1. Resistivity ρ and saturation magnetization M_s of 1000 Å thick $Co_{1-x}Ge_x$ (x = 0.42, 0.50, 0.58, 0.81, 1.00) single layers.

Spacer	ρ ($\mu\Omega$ cm)	M_s(G)
Ge	>20000	0
$Co_{0.19}Ge_{0.81}$	18000	0
$Co_{0.42}Ge_{0.58}$	2000	0
$Co_{0.50}Ge_{0.50}$	485	0
$Co_{0.58}Ge_{0.42}$	362	0

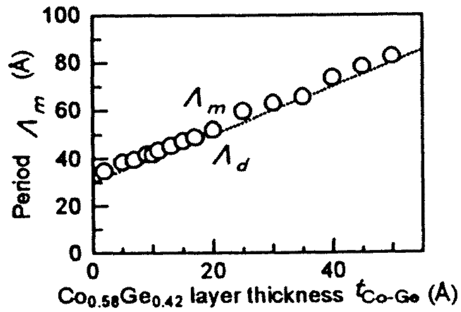

Figure 3. Superlattice period Λ_m of [Co(30 Å)/$Co_{0.42}Ge_{0.58}$(t_{Co-Ge} Å)]$_{22}$ as a function of the spacer thickness. The dotted line shows the designed period Λ_d.

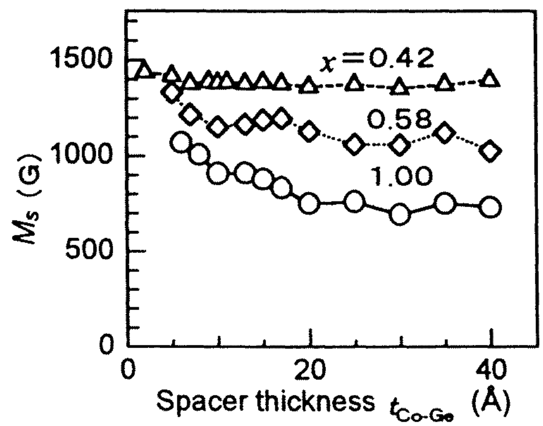

Figure 4. Saturation magnetization M_s of [Co(30 Å)/$Co_{1-x}Ge_x$(t_{Co-Ge} Å)]$_{22}$ superlattices ($x = 0.42, 0.58, 1.00$) as functions of the spacer thickness. Empty circles (○), diamonds (◇) and triangles (△) show the values for Ge, $Co_{0.42}$ $Ge_{0.58}$ and $Co_{0.58}$ $Ge_{0.42}$, respectively.

Ge_x (t_{Co-Ge} Å)]$_{22}$ ($x = 0.42, 0.50, 0.58, 0.81, 1.00$) superlattices. Precise XRD analyses have revealed that interdiffusion at the interfaces is considerably suppressed by increasing the Co content. Figure 3 shows the measured superlattice period Λ_m and the designed Λ_d (dotted line in the figure) against t_{Co-Ge} for $x = 0.42$. For the whole thickness range, Λ_m perfectly coincides with Λ_d, indicating very little interdiffusion at the interfaces. The absence of interdiffusion for this composition is also supported by the fact that the samples maintain constant M_s equal to that of pure Co(1420G), as indicated by (△) in figure 4. Figure 5 shows the coupling behaviours as a function of the spacer thickness of Co/$Co_{1-x}Ge_x$ superlattices. For all the spacer compositions in the range $0.42 \leqslant x \leqslant 0.81$, one can find evidence of AF coupling similar to Co/Ge superlattices. FMR measurements have also revealed that the coupling oscillates from F to AF around $t_{Co-Ge} = 10$ Å and then goes to non-coupling. These results clearly demonstrate that a non-magnetic Co–Ge spacer can mediate AF coupling between adjacent Co layers, which is responsible for the AF coupling found in Co/Ge superlattices. This situation is very similar to Fe/Si superlattices where Fe-Si diffused regions contribute to strong AF coupling [3].

In summary, we first found evidence of AF coupling in Co/Ge superlattices and the

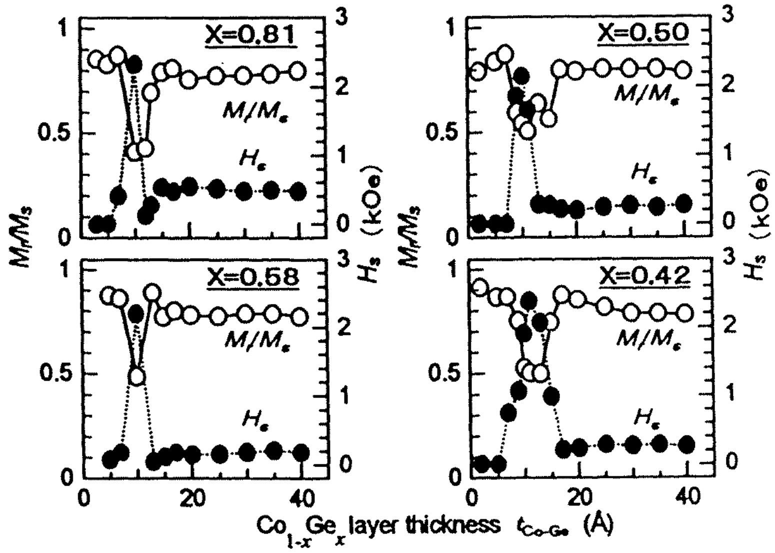

Figure 5. Variations of the remanence ratio M_r/M_s (○) and the saturation field H_s (●) of $[\mathrm{Co}(30\ \text{Å})/\mathrm{Co}_{1-x}\mathrm{Ge}_x(t_{\mathrm{Ge}}\ \text{Å})]_{22}$ ($x = 0.42, 0.50, 0.58, 0.81$) superlattices against the spacer thickness.

coupling strength was about 50 merg cm^{-2}, much smaller than that of Fe/Si superlattices. Precise structural analyses revealed that this AF coupling was mediated by non-magnetic Co–Ge diffused substance. This result was also supported by the fact that similar AF coupling was unambiguously observed in Co/non-magnetic Co–Ge superlattices.

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